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Two-Photon Single-Beam “Bit-by-Bit” Writing and Fluorescent “Page-by-Page” Reading 2.5D Femtosecond Optical Data Storage System

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A novel scheme of rewritable multilayered (2.5D) optical data storage based on single-beam femtosecond two-photon consequent writing and fluorescent parallel readout has been developed. The table-top prototype is created to demonstrate writing-reading-erasing the information in dual-layer polymer photochromic matrices with equivalent information density of 50 Mbit/cm² per layer.

KEYWORDS: 3-dimensional optical data storage, femtosecond, two-photon absorption, photoisomerization

Three-dimensional optical memory is a promising technology for increasing the capacity of computer data storage. Several physical principles such as holography, two-photon absorption photochemistry, hole burning, photoinduced poling, optical damage and photon echo have been suggested as methods to use in the development of 3-D optical data storage technology. One prospective approach is connected with the use of organic photochromic molecules embedded in polymer matrices.^{1,2} A variant based on two-photon two-beams picosecond “page-by-page” writing and fluorescent “page-by-page” reading has been successfully developed.³

Previously we reported some results on two-photon data writing.⁴ Two beams of picosecond pulsed radiation at different wavelengths are used. Here we present a novel variant of a quasi-3D multilayered optical memory based on single-beam two-photon femtosecond “bit-by-bit” writing with the assistance of continuous trains of femtosecond pulses and fluorescent “page-by-page” reading.

Multilayer structures with alternating photoactive and non-photoactive polymer layers are used as data storage media. (Fig. 1) Localization of volumetric information pixels (voxels) inside the storage medium is provided by the non-linearity of two-photon writing and by the thin photosensitive layers. We will refer to such multilayer optical memory media as 2.5-dimensional matrices.

Application of a single-beam writing principle in 2.5-dimensional optical memory devices enables us to use very short femtosecond laser pulses, and therefore, to significantly decrease the “energy cost” of the information bit recording. A prototype based on single-beam two-photon successive writing and “page-by-page” fluorescent parallel reading with single-photon excitation has been created. The scheme of the prototype is depicted in Fig. 2.

The writing process is the photocoloration of naphthacenopyridones (NP), i.e. the transformation of NP from form A into form B.⁵ Femtosecond pulses generated by a Ti:sapph. self-modelocked cw laser pumped by an argon-ion laser were used to provide writing in 2.5D matrices with two photoactive layers sensitized by NP. Writing wavelength is tuned in the range of 800–860 nm. The pulse repetition rate is 100 MHz. Average power of the writing beam at the storage media did not exceed 50 mW while pulse duration was ~200 fs. Writing exposure is changed by varying the number of pulses in trains

from 10³ to 10⁶ with the help of the electrooptical gate. Inside the universal head writing radiation is reflected by a dichroic mirror into a microscopic objective (×40, N.A. 0.65) that focuses this radiation into a 1.5-μm spot. The optical storage medium is mounted on precision 3D stage driven by step motors.

Flat pages of information are read by exciting the fluorescence of colored form of photochromic molecules with the help of a portion of cw argon-ion laser radiation. The reading wavelength is 514.5 nm, the average power of the readout beam is ~100 μW and the beam diameter is approximately 1 mm. Exposure is determined by the acoustooptic gate. The reading beam is spatially formed to provide the uniform lighting of 110 μm × 160 μm pages disposed in different layers of the 2.5D matrix. Fluorescent images are magnified using the same microscopic objective that is used to focus the writing beam. After spectral selection, the images are registered using CCD camera “Princeton Instruments MicroMax”. To select the appropriate layer, the microscopic objective is refocused. The computer controls the gates, 3D stage and CCD

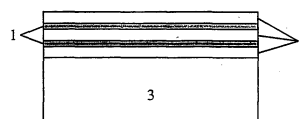


Fig. 1. Cross section of multilayer optical data storage medium (2.5D optical matrix). 1 – photosensitive layers (~1 μm thickness); 2 – separating non-photoactive polymer layers (30 μm thickness); 3 – glass substrate.

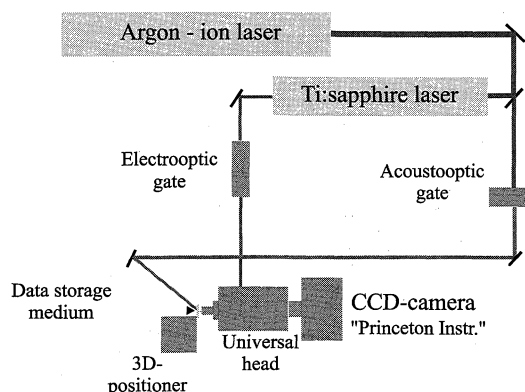


Fig. 2. The scheme of the 2.5D optical data storage prototype.

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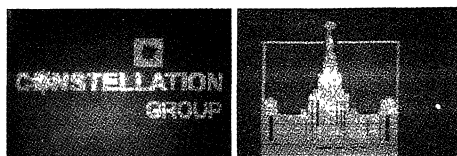


Fig. 3. Fluorescent microscopic images ($160 \times 110 \mu\text{m}$) of two neighbor layers containing 0.2 M of NP in methyl methacrylate-styrene-acrylonitrile copolymer.

camera.

As an example, two images from the different layers of the 2.5D matrix are shown in Fig. 3. The position of the first image is directly in front of the second one. In spite of rather high data density in the images, only a weak background is registered.

To give the simplest analytical description of the recording process, let us assume the following:

- interactions between photochromic molecules and the environment dose not effect the process;
- initially, all photochromic molecules are in form A;
- writing light interacts only with photochromic molecules in form A and only the first electronic level is excited;
- the photochemical destruction of molecules and reverse reaction are negligible, i.e., their total number in both isomer forms is constant;
- the polymeric medium containing photochromic molecules is homogeneous.

For femtosecond pulse trains, the relaxation time of exited state of NP is much larger than the pulse duration, but smaller than the period between pulses.

Under these assumptions, the accumulation of colored form after propagation of k femtosecond light pulses can be described as

$$n_B(k) = n_0 \left\{ 1 - \left[1 - \frac{\varphi}{2} \left(1 - \exp(-2\delta g \frac{E^2}{\tau}) \right) \right]^k \right\} \quad (1)$$

where $n_B(k)$ is the concentration of colored form, n_0 is the total concentration of photochromic molecules, φ is the coloration quantum yield, δ is the two-photon absorption cross section, g is the form-factor of a femtosecond laser pulse, E is the density of light pulse energy and τ is the pulse duration.

One can see that if the effective cross section in the form $\sigma_{\text{eff}} = \delta g E / \tau$ is used the action of light may be described by a formula similar to that used for the description of a single-photon process.

Here we introduce the reduced energetic cost (REC) of single informative bit recording:

$$REC = kE \sqrt{\frac{\delta g}{\tau}} \quad (2)$$

According to eq. (1) REC depends on the number of pulses as

$$REC = k \sqrt{\frac{1}{2} \ln \left\{ 1 - \frac{2}{\varphi} \left[1 - \left(1 - \frac{n_B}{n_0} \right)^{\frac{1}{k}} \right] \right\}^{-1}} \quad (3)$$

This formula allows an optimal number of pulses for any fixed concentration of colored form to be determined. For NP used in the prototype, this number is close to 10.

The information readout is accompanied by partial erasing due to photobleaching of photochromic molecules in colored form.

It is obvious that the maximum number of readout cycles is limited by

$$N_{\text{max}} = \left[\frac{\varphi n n_B(0)}{\varphi_B \Delta F_{\text{min}}} \right] = \left[\frac{F_0}{\Delta F_{\text{min}}} \right], \quad (4)$$

where ΔF_{min} is the minimal quantity of fluorescent photons required for their registration. Square brackets indicate the integer part.

If the reading of information is performed using equal doses of illumination in every readout cycle, and $N_{\text{max}} \gg 1$ the maximum number of readout cycles and the optimal pulse energy of reading E^{opt} are defined as

$$K_{\text{max}} = \left[\frac{N_{\text{max}}}{e} \right] + 1 \quad (5)$$

and $E^{\text{opt}} \cong e / N_{\text{max}}$.

For a more detailed analysis, the computer code has been expanded by taking into account the reversibility of photochemical reactions, the inhomogeneity of photochromic molecules embedded into polymer host, the photodestruction and the energy transfer in highly doped photochromic polymeric media.

A prototype of a 2.5D optical data storage device has been developed. Single-beam femtosecond two-photon writing and fluorescent reading as well as single-photon excited erasing have been demonstrated. Test images have been recorded into the single- and two-layered polymer photochromic matrices sensitized by NP derivatives. The measured value of two-photon cross section of NP No. 10⁵) in form A at a wavelength of 860 nm is proved to be about $10^{-51} \text{ cm}^4\text{s}$.

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